Diffusion at low-temperature moisturization and high-temperature cooking of wheat

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Abstract. A review has been made of studies in diffusion coefficients values and their dependence on temperature of wheat moisturization (whole grain, scoured grain, grain kernel, endosperm, and pericarp). Experimental research was performed at temperatures between 10 ºC and 98.3 ºC. Values of diffusion coefficients were found by using approximated solutions to the unsteady-state diffusion equation. The values of diffusion coefficients vary over a wide range between $1.1 \cdot 10^{-12}$ m^2/sec (at 10 ºC) and $1.4 \cdot 10^{-10}$ m^2/sec (at 98.3 ºC). The relation between the diffusion coefficient and the temperature is described by the Arrhenius equation. Studies in this work were performed at higher temperatures between 105 and 140 ºC. At these temperatures, wheat is cooked, and in order to determine diffusion coefficients one has to use solution of unsteady-state diffusion by the numerical grid method in an implicit scheme. This solution made it possible to take into account changing diffusion coefficient with changing temperature, as well as separation of the outer starch layers and reduction of material particles size to complete cooking. In order to compare calculation results obtained using grids method and the steady-state conditions approximate equation that is used by authors of previous works, we performed analytical solution of transient diffusion equation for an infinite plate using the method of variable separation. It has been shown that calculation results of the analytical solution are consistent with results of the grid method and are different from the results for regular mode only during the initial period. The greatest computer time consumption corresponds to analytical solution. It has been shown that diffusion coefficients during cooking can only be defined when a mathematical model is used, which is based on numerical method of solving unsteady diffusion equations. It has been established that during cooking, diffusion coefficient is substantially lower than that in case of material moisturization, and is equal to $8.273 \cdot 10^{-12}$m^2/s at 138 ºC. This can be explained by the fact that changes in starch granules occur in course of cooking.

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Introduction

In the process of drying grain and products of its processing, in course of moisturization in production of flour and in course of corn grits cooking in alcohol production, processes of moisture diffusion occur. For the purpose of modeling these processes, knowledge of diffusion coefficients is required. Diffusion coefficients are determined experimentally in course of drying and moisturization, by measuring weight of material over time. This article examines the diffusion coefficients obtained by wheat moisturization and cooking. Diffusion coefficients after moisturization have been determined for relatively low temperatures. Practically, in this range they obey the Arrhenius equation and have little dependency on the shape of particles and the type of raw material. Experimental data are processed in an unsteady-state diffusion equation for regular geometric shapes (infinite plate, infinite cylinder, sphere) that has been simplified and solved analytically for the regular mode with the following initial and boundary conditions [1]:

1. Initially, moisture concentration in the material is distributed uniformly.
2. Resistance to mass transfer in the liquid phase is negligible and concentration of moisture on the surface of a solid is balanced with the surrounding liquid.
3. For any point in time $\tau > 0$, concentration of moisture on the surface of the material is constant.

General solution for an infinite plate, infinite cylinder and sphere is as follows:

$$\frac{\overline{m} - m_0}{m^* - m_0} = 1 - \sum_{n=1}^{\infty} B_n \exp\left(-A_n^2 \mathrm{Fo}\right), \quad (1)$$

where $\overline{m}$ is the average humidity over time, % of dry basis; m^* is the equilibrium moisture on the surface of the kernel, % of dry basis; m_0 is the initial moisture content in the entire region of the kernel, % of dry basis; $A_n = const$ [$(2n-1)(\pi/2)$ for an infinite plate; $r_c\alpha_n$ for an infinite cylinder and $n\pi$ for sphere]; $B_n = const$ [$2/A_n^2$ for an infinite plate; $4/A_n^2$ for an infinite cylinder and $6/A_n^2$ forsphere]; Fo is the Fourier number $\mathrm{Fo} = D\tau/\mathrm{L}_\mathrm{c}^2$; D is the diffusion coefficient, m²/s; $\mathrm{L_c}$ is the geometric parameter, m [the half-thickness of plate - for an infinite plate; cylinder radius - for an infinite cylinder; ball radius - for ball]; $r_c\alpha_n$ is the n-th positive root of the Bessel function $J_0(r_c\alpha_n)=0$; $J_0(x)$ is the zero-order Bessel function of the first kind; τ is time, s.

Influence of temperature is considered in the Arrhenius equation

$$D = D_0 e^{-E_a/RT}, \quad (2)$$

where D_0 is the pre-exponential factor (diffusion factor); E_a is activation energy, J/mol; T is the absolute temperature, K; R is gas constant, R = 8.314 J/(mol·K).

The pre-exponential multiplier D_0 and the activation energy E_a were calculated from the linear regression in representation of the diffusion coefficient natural logarithm as a function of a value inverse to the absolute temperature.

Materials and methods

We studied the diffusion coefficient at 105-138 ºC, at which temperature the wheat corn middlings were cooked.

In course of cooking, processes of moisturization, swelling of the starch granules, changing temperature and peeling of the outer layer of starch granules occur. The middlings were cooked after roll mills, and were considered as an infinite plate. Basing on the analysis of starch granules swelling rate performed by various researchers and generalized in work [2], it was found that the swelling rate is significantly higher than the diffusion rate. Therefore, we considered diffusion a limiting stage, and found diffusion coefficient in the process of cooking. Peeling of the outer layer of particles is also caused by moisture delivery into the material by diffusion. Due to the complexity of the process, it can only be described on the basis of numerical solution. For checking accuracy of the numerical solution method and the assumptions of the regular mode used by many authors, we presented an analytical solution of an unsteady-state diffusion equation for a flat plate.

Let's consider moisturization of a particle with thickness δ, initial humidity C_0 and the equilibrium moisture content bordering with water C^*. In the particle, unsteady-state diffusion occurs.

$$\frac{\partial C}{\partial \tau} = D\frac{\partial^2 C}{\partial x^2}. \quad (3)$$

Initial conditions

$$C(x,0) = C_0. \quad (4)$$

Boundary conditions

$$C\left(\frac{\delta}{2},\tau\right) = C^*. \quad (5)$$

$$\left(\frac{\partial C}{\partial x}\right)_{x=0} = 0, \quad (6)$$

where x is the coordinate, m.

Equation (1) is converted to (7)

$$\frac{\partial C}{\partial \tau} = \theta^2 \frac{\partial^2 C}{\partial \psi^2} \quad (7)$$

where

$$\psi = \frac{x}{\delta/2}; \quad \theta^2 = \frac{D}{\left(\delta/2\right)^2} \quad (8)$$

Let us state $y = C - C^*$. Let us use the variable separation method. Let us represent the solution as

$$y(\psi,\tau) = X(\psi)\cdot T(\tau) \quad (9)$$

After transformations, we get

$$\frac{1}{\theta^2}\frac{1}{T(\tau)}\frac{\partial T}{\partial \tau} = -\lambda^2 \quad (10)$$

$$\frac{1}{X(\psi)}\frac{\partial^2 X}{\partial \psi^2} = -\lambda^2 \quad (11)$$

Solution of equation (10) is

$$\frac{1}{T(\tau)}\frac{\partial T}{\partial \tau} = -\lambda^2\theta^2 \quad (12)$$

$$T(\tau) = \exp\left(-\lambda^2\theta^2\tau\right) \qquad (13)$$

Solution of equation (11) is

$$X(\psi) = A\sin(\lambda\psi) + B\cos(\lambda\psi) \qquad (14)$$

Taking into account boundary conditions, we find:

$$y(\psi,\tau) = \sum_{n=0}^{\infty}\left[B_n \cos\left(\frac{2n+1}{2}\pi\psi\right)\cdot\exp\left(-\frac{(2n+1)^2}{4}\pi^2\theta^2\tau\right)\right] \qquad (15)$$

where

$$B_n = \frac{4\left(C_0 - C^*\right)}{(2n+1)\pi}\sin\left(\frac{(2n+1)}{2}\pi\right) \qquad (16)$$

Inserting result (16) into equation (15), we obtain

$$y(\psi,\tau) = \sum_{n=0}^{\infty}\left[\frac{4\left(C_0 - C^*\right)}{(2n+1)\pi}\sin\left(\frac{(2n+1)}{2}\pi\right)\cos\left(\frac{2n+1}{2}\pi\psi\right)\cdot\exp\left(-\frac{(2n+1)^2}{4}\pi^2\theta^2\tau\right)\right] \qquad (17)$$

Given that under the sign of the sum, value $\sin\left(\frac{(2n+1)}{2}\pi\right)$ alternatively takes values -1 and +1, we can write

$$y(\psi,\tau) = \sum_{n=0}^{\infty}\left[\frac{4\left(C_0 - C^*\right)}{(2n+1)\pi}(-1)^n\cos\left(\frac{2n+1}{2}\pi\psi\right)\cdot\exp\left(-\frac{(2n+1)^2}{4}\pi^2\theta^2\tau\right)\right] \qquad (18)$$

Returning from y to C, taking into account the introduced variable $y = C - C^*$ substitution, taking the constant $\left(C_0 - C^*\right)$ outside the summation sign, we finally get

$$\frac{C - C^*}{C_0 - C^*} = \sum_{n=0}^{\infty}\left[\frac{4}{(2n+1)\pi}(-1)^n\exp\left(-\frac{D}{\left(\delta/2\right)^2}\frac{(2n+1)^2}{4}\pi^2\tau\right)\cos\left(\frac{2n+1}{2}\pi\frac{x}{\delta/2}\right)\right] \qquad (19)$$

The above equation (19) is reduced to equation (1) when averaged over the coordinate in regular mode.

We have studied these processes by analyzing the images under a microscope. By measuring dimensions of initial granule and a granule swollen to the size at which the starch granules burst, it was established that at concentration of 80 % wt., the outer layer peels off. A mathematical model has been developed. The solution was made using the grid method. The solution takes into account kinetics of swelling described by the equation of chemical reaction of 2-nd kind [3].

$$\frac{dX}{d\tau} = V(\tau)(1 - X)^2, \qquad (20)$$

where $V(\tau)$ is the rate constant that depends on temperature and is calculated according to the Arrhenius equation.

$$X = \frac{D_t - D_0}{D_m - D_0}, \qquad (21)$$

where D_0 is the diameter of initial (dry) starch granule, microns; D_m is the maximum diameter of swollen starch granule, microns; D_t is current diameter of a starch granule (swelling) microns.

We also have taken into account the decrease in size of a cooked particle in course of outer layer peeling, change of boundary conditions, and influence of temperature. Model identification is based on the data about wheat middlings cooking sequentially at temperatures 138 and 105 ºC for 60 and 20 minutes. In course of the identification process, the value of pre-exponential factor was changed, and the value of activation energy was chosen, at which complete cooking occurred in the above conditions. Figure 1 shows the change in moisture concentration in various coordinate layers. The total number of layers was 100. On the last 100-th layer, complete particles cooking occurred. For 60 minutes the temperature was maintained at 138 ºC. After that, the temperature was sharply changed to 105 ºC, therefore, a sharp decrease is observed in the concentration graphs on the 60-th minute. The basic value of the activation energy is equal to 32.12 kJ/mol, which is closest to the data presented in the literature. The curves that correspond to it are shown in Figure 1 by solid lines. For 138 and 105 ºC modes, equal values have been taken- D_0 and E_a. This is caused by the fact that the mode with temperature 138 ºC continued for 60 minutes. Solid substances of plant origin undergo substantial changes in structure with temperature increasing. During this time, major structural changes in particles occurred that persisted for next 20 minutes at 105 ºC. The diffusion coefficient in this case was $3.641\cdot10^{-12}$m²/s.

Strictly speaking, the grain, unlike solid inorganic material, is a multicomponent, containing starch, proteins, fats and carbohydrates. Moreover, these substances are essentially polymers. The structure of these polymers changes in course of heating and moisturization. For this reason, it is impossible to use the diffusion equation and mass transfer for describing processes in multicomponent mixtures [4].

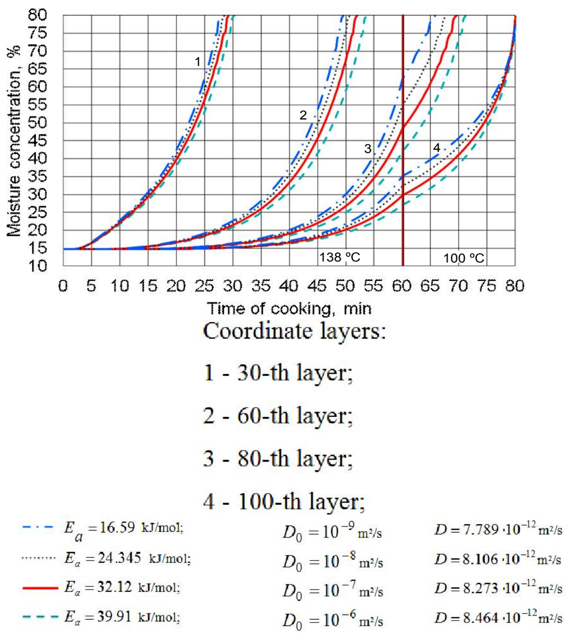
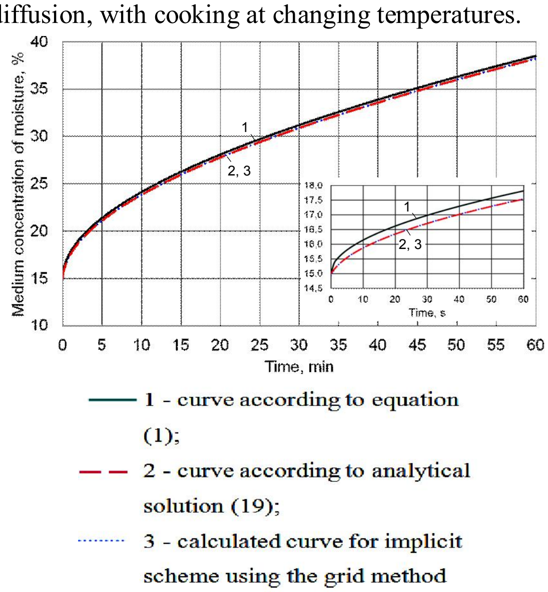


Figure 1. Change in moisture concentration with time in coordinate layers

Using the above exact unstable-state diffusion solution (19), we assessed accuracy of the used cooking model based on numerical solution using grid method for an implicit scheme, and on diffusion equation in regular mode (1). The results of the solution are shown in Figure 2, which shows that description of diffusion at 138 ºC coincides with the data of numerical solution with use of the grid method (curves 2 and 3). The calculation using equation (1) (curve 1) deviates from the exact solution only during the initial interval. This is the basis for comparing all results using isothermal diffusion, with cooking at changing temperatures.



The diffusion coefficient in grains kernel was studied at relatively low process temperatures. This is due to the fact that in the temperature range between 20 and 70 ºC, the structure of the material does not undergo major changes and, as shown by experiment, data about diffusion coefficients are satisfactorily described by the Arrhenius equation. The results indicate that, in accordance with the Arrhenius equation, the diffusion coefficient increases with temperature. In industrial processes, the processes of cereal middlings saturation with moisture occur in case of cooking at higher temperatures as well, up to 155 ºC. It did not seem feasible to obtain experimental data about diffusion coefficients at these temperatures, since irreversible changes in the structure of grain materials occurred during cooking. Meanwhile, extensive data have been obtained about grains cooking at enterprises of the alcohol industry.

Discussion

Let us compare the obtained data about diffusion coefficient to the data published for various wheat grades

For all wheat grades, the values of endosperm diffusion coefficient were in the range between $0.46 \cdot 10^{-10}$ and $1.4 \cdot 10^{-10}$ m²/s (Table 1). Pericarp diffusion coefficients are in the range between $0.042 \cdot 10^{-10}$ and $0.42 \cdot 10^{-10}$ m²/s (Table 1) [5]. The endosperm diffusion coefficients are more than that of pericarp.

Table 1. Wheat endosperm and pericarp diffusion coefficients

Grade	Diffusion coefficient, m²/s at 22 ºC. $D \cdot 10^{10}$	
	Endosperm	Pericarp
Grandin	0.8	0.13
Amidon	0.55	0.16
Renville	0.73	0.13
Jagger	0.91	0.13
TAM107	1.4	0.042
Madsen	0.6	0.19
Rely	0.46	0.32
Penawawa	0.55	0.42
Vanna	0.57	0.29

As compared to other studies, these values are lower than those obtained using NMR [6, 7]. However, other researchers [8-11] report lower diffusion coefficients (Table2).

Table 2. Wheat diffusion coefficient

Source	Wheat grade	Temperature, ºC	Coefficient of diffusion, $D \cdot 10^{10}$ m²/s
6	Aotea (endosperm)	22	1.8
7	Otane	22	12
8	Thatcher	20.8-79.5	0.069-7.2
9	Thatcher	25	0.018-0.031
10	Ponca Vernum Seneca Brevor	26.7-98.3 30-86 26.7-98.3 30-86	0.027-2.456 0.022-0.752 0.031-1.409 0.027-0.891
11	Wheat kernel	10 50	0.011 0.1

In work [12], the effective water diffusion coefficient during wheat kernel moisturization varied between $2.80 \cdot 10^{-12}$ and $1.36 \cdot 10^{-11}$ m²/s with activation energy of 34.26 kJ/mol with temperature ranging from 25 to 65 ºC (Table 3).

Table 3. Effective wheat kernel diffusion coefficient at various moisturization temperatures

Temperature, ºC	Effective diffusion coefficient, m²/s
25	$2.8 \cdot 10^{-12}$
35	$4.18 \cdot 10^{-12}$
45	$6.24 \cdot 10^{-12}$
55	$1.06 \cdot 10^{-11}$
65	$1.36 \cdot 10^{-11}$

Work [13] shows diffusion coefficients of hulled and whole wheat grains for various geometries at 22 ºC (Table 4).

Table 4. Diffusion coefficient for hulled and whole wheat grains

Grade	Diffusion coefficient $D \cdot 10^{10}$, m²/s			
	for hulled grains		for whole grains	
	ball	ellipsoid	ball	ellipsoid
Grandin	0.62	0.60	0.18	0.16
Amidon	0.41	0.39	0.17	0.14
Renville	0.55	0.48	0.17	0.13
Jagger	0.70	0.67	0.19	0.16
TAM107	1.09	1.04	0.05	0.04
Madsen	0.48	0.42	0.20	0.17
Rely	0.47	0.46	0.23	0.19
Penawawa	0.44	0.44	0.29	0.25
Vanna	0.44	0.43	0.25	0.21

For all wheat grades, diffusion coefficient values calculated for endosperm ranged between $0.39 \cdot 10^{-10}$ and $1.04 \cdot 10^{-10}$ m²/s. Values of moisture diffusion coefficient for whole wheat kernel were in the range between $0.04 \cdot 10^{-10}$ and $0.29 \cdot 10^{-10}$ m²/s.

The above experimental data about diffusion coefficients are shown in Figure 3 as function Ln (D) of temperature.

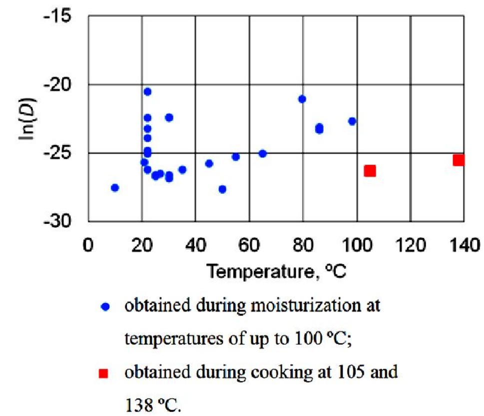


Figure 3. Dependence of $\ln(D)$ on temperature

High values of diffusion coefficients at 22º C were obtained by NMR method (Table 2) and for endosperm (Table 1). If these data are not considered, it can be seen that in the temperature range between 20 and 100º C, diffusion coefficients increase with increase in temperature, practically obeying the Arrhenius equation. With further increase in temperature up to 140 ºC, diffusion coefficient decreases. As shown above, the reason for such decrease is structural change in starch granules at high temperatures.

Conclusion

In modeling the cooking process in the range between 105 and 140 ºC, for calculating the diffusion coefficient it is recommended to use equation (2) with constants $E_a = 32.12$ kJ/mol and $D_0 = 10^{-7}$ m²/s.

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